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Accelerated Creep Mechanics: Parts I and II

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ACCELERATED CREEP MECHANICS: Part I

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ABSTRACT

At IPST, we feel we have made progress in understanding accelerated creep. We have taken ideas expressed in the past, combined them with our own thoughts, and proposed a mechanism for accelerated creep. We believe that accelerated creep is a natural consequence of cyclic stress gradients and certain characteristics of the creep constitutive behavior for the material. Cyclic stress gradients arise during cyclic humidity testing as a result of moisture gradients and/or inhomogeneous material properties. The pertinent constitutive behavior is that the material will creep more under cyclic load than at constant mean load.

We express our mechanism in mathematical form and provide numerical simulations of accelerated creep in paper. We demonstrate that the model mimics the creep behavior of paper and argue that accelerated creep is a response to the creep constitutive behavior. Unlike other theories, our explanation does not require the introduction of new material concepts or new phenomena related to sorption.

INTRODUCTION

In this paper we attempt to put forth a plausible explanation of the accelerated creep phenomenon. According to our model, accelerated creep should be the expected outcome of a cyclic humidity creep test for a certain class of materials. Our simple model does not invoke any extraordinary assumptions such as imposed dependence on rate of moisture change, or special structural features of fibers and paper. Our assumptions for material behavior are well founded for polymers as well as other materials, and our prediction of accelerated creep is based on first principles.

There is no doubt that our mechanism yields accelerated creep, and as long as the conditions we set forth are met accelerated creep will be active. As will be demonstrated in this paper, experimental observations of accelerated creep in polymers conform to the response predicted by our model. We still must demonstrate that our mechanism of accelerated creep is sufficient to explain accelerated creep in paper. This paper and the following paper, Accelerated Creep Mechanics: Part II, provide strong support for our mechanism.

First, we must define what we mean by accelerated creep. For us, accelerated creep occurs when the effective creep rate in cyclic humidity is higher than the creep rate at high humidity. Many polymers, including paper, exhibit a secondary creep response that is linear with a log scale of time. As shown in Figure 1, we define the accelerated creep ratio as the ratio of the slopes of the creep versus log time for the cyclic RH region and the constant high RH region.

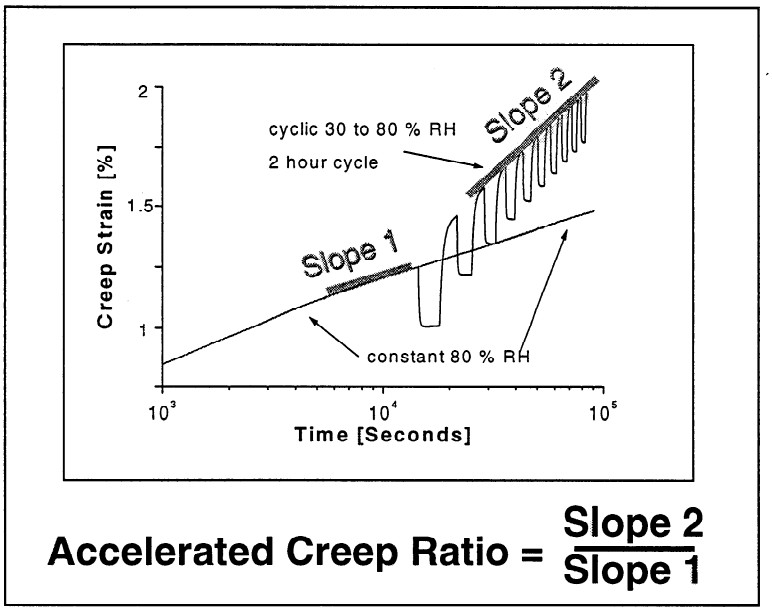

Figure 1. Accelerated creep ratio.

MECHANISM OF ACCELERATED CREEP

Our criterion for accelerated creep is that the creep strain created in a sample subjected to one cycle of high and low moisture is greater than the creep strain created during the same time interval for a sample maintained at high moisture content. In order to understand accelerated creep, we need to express this criterion in terms of the requirements on the physical state and material behavior that will lead to accelerated creep. In the following, we describe the development of our mechanism and point out any assumptions that we are making along the way.

To begin, we propose that accelerated creep is the expected result of a creep test in cyclic humidity for a certain class of materials. First, we assume that such a material must be sensitive to moisture. For example it may be hygroexpansive and/or possess moisture dependent material properties. Such is the case for a variety of polymers. Second, we assume that the material is viscoelastic-plastic and will creep under a sustained load. At this point, we will not place any further limitations on the material behavior, but rely on

the criterion for accelerated creep given above to impose any restrictions on the constitutive behavior.

In order to have a physical picture in mind, consider the tensile test shown in Figure 2. For a prescribed environment, the total strain, $\varepsilon(t)$, is defined as the change in length divided by the total length and is a function of time, t, only. In other words, we are assuming uniform strain. The total applied force, P, is constant, but the axial stress, $\sigma(z,t)$ may be a function of time, t, and position, z.

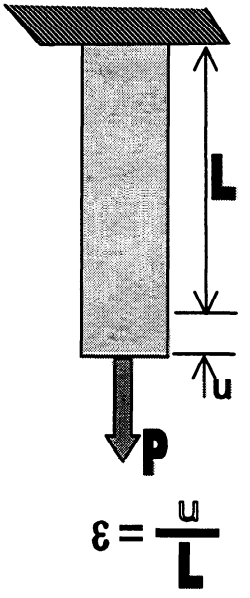

Figure 2. Tensile creep test.

Assume that at any given time, the total strain is comprised of a linear elastic component, a creep component, and a linear hygroexpansive component. We write the strain rate as

$$\frac{d\varepsilon(t)}{dt} = \frac{1}{E(z,m)}\frac{\partial\sigma(z,t)}{\partial t} + F[\sigma,\varepsilon,m,z;t] + \beta(z,m)\frac{\partial m(z,t)}{\partial t} \qquad (1)$$

where E is the elastic modulus, β is the coefficient of hygroexpansion, and m is the moisture content. The creep term, F, will be, in general, a functional that depends on the entire history of the material.

Because the strain is not a function of position, Equation (1) shows that the stress most likely will be a function of position across the thickness (could be the width) of the sample if either the moisture or the mechanical properties are functions of position. If the moisture enters the material by diffusion, we may expect moisture gradients in the specimen. The moisture gradients, in turn, lead to nonuniform changes in the potential for hygroexpansion. This will likely lead to nonuniform stresses. In a similar manner, if we have heterogeneity and/or moisture dependence of properties such as $\beta(z,m)$ and $E(z,m)$, then the stresses most likely will not be uniformly distributed.

If the change in moisture triggers a change in the stress distribution, then every time we change the moisture, the stress distribution in the sample will change. Figure 3 illustrates the link between the stress distribution and moisture gradients.

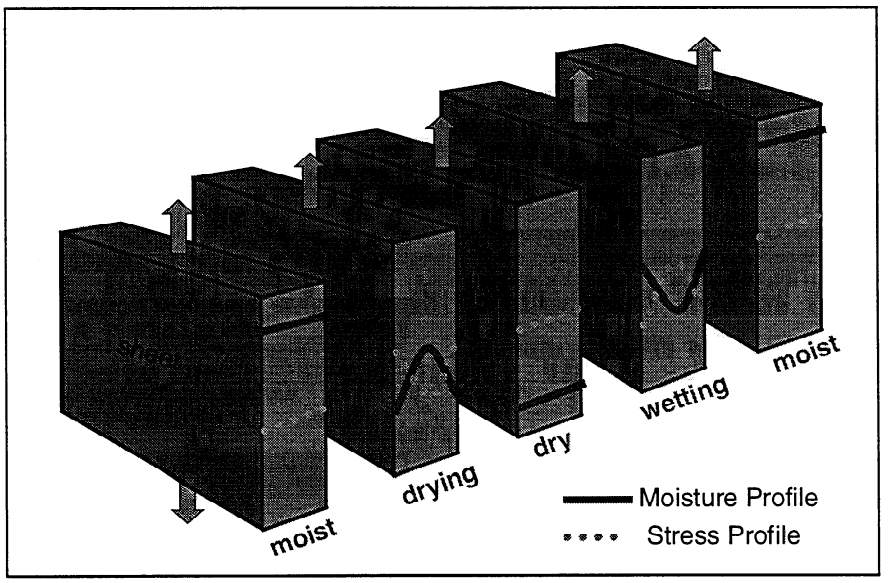

Figure 3. Moisture gradients lead to stress gradients.

Let us start at high moisture and a uniform stress distribution. As we start to dry the sample, the moisture content on the outer edges will be lower than the center. The differential moisture change creates a larger potential for shrinkage on the outer edges, but the sample is constrained to have uniform strain. This constraint leads to additional tension on the edges and lowered tension in the middle. After the moisture level reaches equilibrium at the low level, and sufficient time has elapsed for relaxation to occur, the stresses will again be uniformly distributed. Now if we re-wet the sample, the outer edges will be under decreased tension and the middle will be under increased tension due to the mismatch in potential for expansion. After we return to equilibrium at the high level of moisture, the stresses will once again move towards a uniform distribution. Thus as we cycle the moisture, each section of the sheet undergoes a cyclic stress. For the case of heterogeneous material properties, the local stresses will cycle as the moisture cycles even without a moisture gradient.

If we integrate Equation (1) over the time required to complete one moisture cycle, the elastic and hygroexpansive components cancel out. We are left with the integral of the creep functional through one cycle. Our criterion for accelerated creep states that the integral of this functional for the case of the cyclic moisture must be greater than the case of constant high moisture. This is written as

$$\int_{cycle} F[\sigma_{cyclic\,RH}, \varepsilon, m_{cyclic\,RH}; t]dt - \int_{cycle} F[\sigma_{high\,RH}, \varepsilon, m_{high\,RH}; t]dt > 0 \qquad (2)$$

One difference in the two integrals of Equation (2) is that the load is variable for the first integral but constant for the second integral. The second difference is that the material properties of the second integral will probably be more compliant, which will need to be overcome by the first integral. For now we focus on the differences in load history. If stress gradients arise from cyclic moisture and the functional that describes the creep behavior of the material satisfies Equation (2), accelerated creep will occur.

We state the requirements for accelerated creep in the following proposed mechanism.

Accelerated creep can occur in materials that exhibit both excess creep under cyclic loading as compared to creep caused by the constant mean load and cyclic redistribution of stresses caused by moisture gradients and/or material heterogeneity.

Our mechanism is not an entirely new idea. In 1942, Pickett [1] investigated accelerated creep of concrete due to cyclic moisture. In his discussion he states:

... an increase in creep accompanying non-uniform shrinkage or swelling is a natural consequence of the fact that the sustained-stress-vs.-strain curve of concrete is not linear.

In the first symposium on moisture effects in board, Selway and Kirkpatrick [2] suggested a similar mechanism for accelerated creep:

Changes in moisture can cause rapid transient increases in stress and as the creep rate is a highly non-linear function of stress there can be significant increases in the creep rate.

These are statements of our mechanism, except in one regard. Whereas both the older statements prescribe the creep as a nonlinear function of stress, we state our material constraint in terms of the cyclic load causing more creep than the mean load. Nonlinear material behavior certainly meets our criteria, but we are specific in ours because we believe that certain materials can yield creep data that appear to be insensitive to load, yet cyclic load causes more creep than the mean load. A Kevlar fiber is an example of such a material.

MECHANICAL MODEL FOR ACCELERATED CREEP

In order to illustrate that our mechanism does indeed yield accelerated creep, we have developed a very simple mechanical model that predicts accelerated creep. This model is also used to demonstrate typical behaviors that should be observed when our mechanism is active.

We need a model that has elastic, creep, and hygroexpansive components. We also must be able to distribute the creep load in at least two parallel components of the structure. Finally, our creep constitutive relation must satisfy Equation (2). The simplest model that incorporates all the necessary components is two Maxwell type elements in parallel as shown in Figure 4.

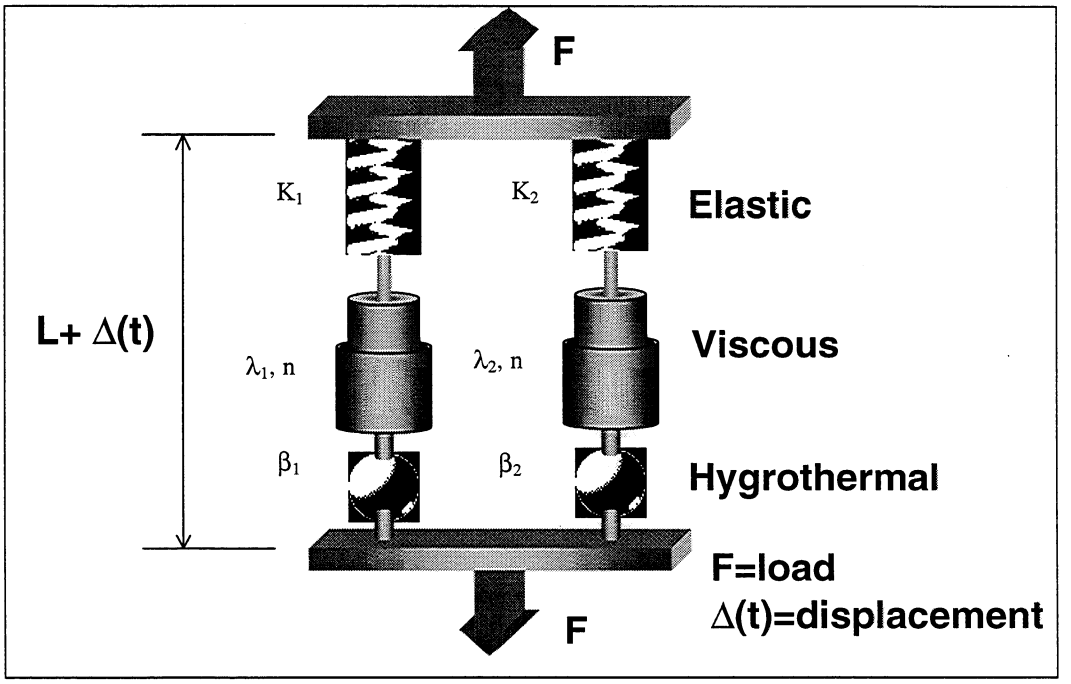

Figure 4. Simple mechanical model to demonstrate accelerated creep.

We will assume the linear elastic springs with stiffnesses, K_1 and K_2; linear hygroexpansion with coefficient of hygroexpansions β_1 and β_2; and dashpots that obey the power law

$$\frac{d\Delta_{dashpot}}{dt} = \lambda_i (F_i)^n;\ i = 1,2 \tag{3}$$

where F_1 is the load carried by Side 1 and F_2 is the load on Side 2 such that

$$F_1 + F_2 = F \ \forall \ t \geq 0. \tag{4}$$

The creep displacement is governed by

$$\frac{d\Delta}{dt} = \frac{1}{K_1}\frac{dF_1}{dt} + \lambda_1 (F_1)^n + \frac{d}{dt}[\beta_1 (m_1(t) - m_1(0))] \quad 0 \leq t < \infty, \tag{5}$$

or equivalently

$$\frac{d\Delta}{dt} = \frac{1}{K_2}\frac{dF_2}{dt} + \lambda_2 (F_2)^n + \frac{d}{dt}[\beta_2 (m_2(t) - m_2(0))], \ 0 \leq t < \infty. \tag{6}$$

Equations (4-6) represent three equations that can be used to solve for the creep displacement, Δ, and the forces F_1 and F_2.

Our criterion for accelerated creep states that the application of a cyclic load causes more creep than the application of the mean load. Let us look at two load cases. Case 1 is the cyclic load

$$F(t) = F_0 + f \sin(\omega t) \tag{7}$$

and Case 2 is the mean load of Case 1 over a period, $t = 2\pi/\omega$,

$$F(t) = F_0. \tag{8}$$

Our condition for excess creep is

$$\int_{cycle} \lambda(F_0 + f\sin(\omega t))^n - F_0{}^n)dt > 0 \tag{9}$$

The inequality (9) is satisfied only if n > 1. For our simple model, excess creep occurs if the dashpot is nonlinear, such that doubling the load more than doubles the creep rate.

Figure 5b shows the excess creep that occurs when n=3 and the load is cycled as shown in Figure 5a. The cyclic loads follow that of Equation (7). For cyclic load 1 the load is first increased, and for cyclic load 2 the load is first decreased. The constant load is the mean load given by Equation (8). Both cyclic creep loads give the same excess creep as compared to the creep resulting from the constant mean load. The creep rate during the time the load is elevated above the mean load causes the excess creep, which is not compensated for by the lower rate of creep when the load is lower than the mean. If n = 1, the total creep from all three load cases would be equal, and if n < 1, the cyclic load conditions would cause less creep than the mean load.

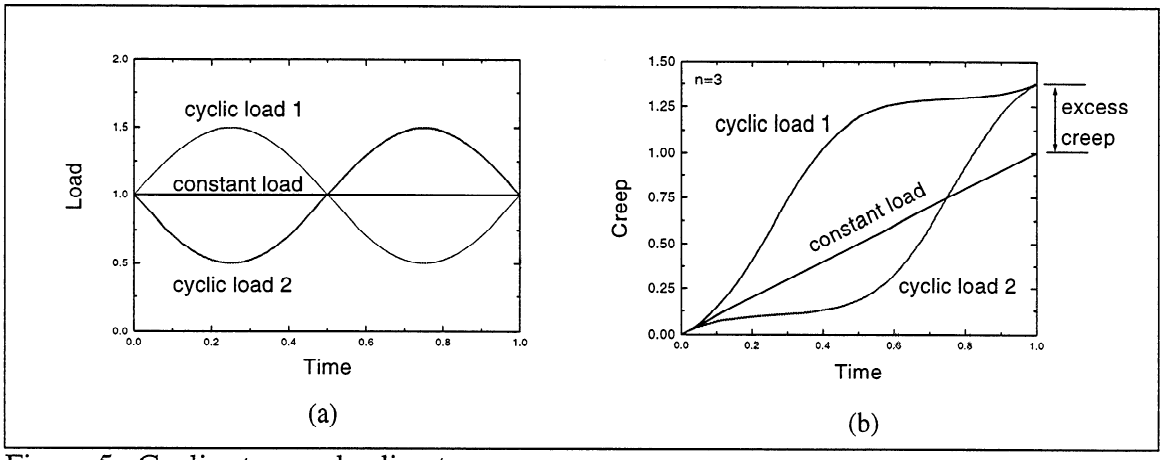

Figure 5. Cyclic stresses leading to excess creep.

In addition to prescribing $n = 3$ we must introduce stress gradients in our model. This is accomplished via our moisture gradients or material heterogeneity. First, let us assume that the moisture in side 2 of our model lags the moisture in side 1 by a time δ/ω. If we set up our accelerated creep experiment by constant high moisture followed by cyclic moisture we can express the moisture contents in each side of our element as

$$m_1(t) = m_0 - \Delta m H(t - t_0)[1 - \cos(\omega t)]$$

and (10)

$$m_2(t) = m_0 - \Delta m H(t - t_0 - \delta/\omega)[1 - \cos(\omega t - \delta)]$$

where $H(t)$ is heavyside step function, m_0 is the wet moisture content, and Δm is extreme change in moisture.

ACCELERATED CREEP

We now have all the elements in place to predict accelerated creep. Consider the case of homogenous material properties that are insensitive to moisture. If we prescribe the moisture gradient given in equation (10) with $\delta = \pi/4$ and the nonlinear dashpot with n = 3, we predict accelerated creep as shown in Figure 6a. Figure 6b shows that there is no accelerated creep for this case if the moisture gradient is removed, $\delta = 0$. The

moisture gradient causes a stress gradient, which combined with the nonlinear creep properties leads to excess creep. Every time we cycle the moisture the stresses are redistributed, which results in accelerated creep. In the case of no gradient, both elements swell and shrink in sync with each other, and the stress is always uniformly distributed. Thus, we do not observe any additional creep.

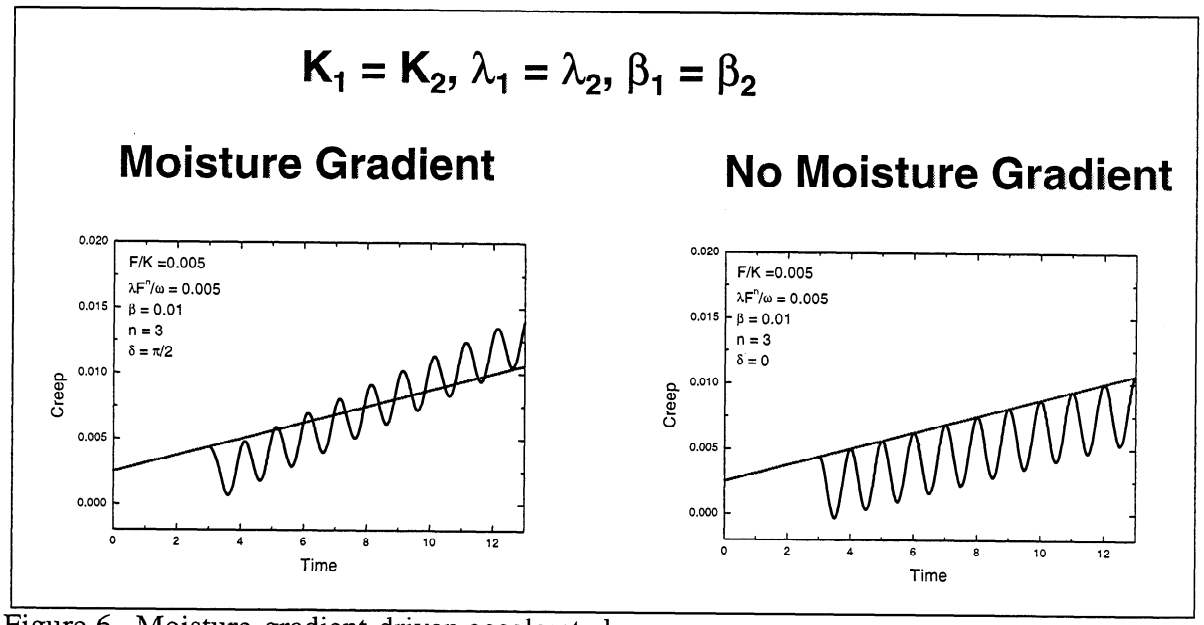

Figure 6. Moisture-gradient-driven accelerated creep.

As previously stated, accelerated creep can arise from heterogeneity in material properties. Figure 7 shows accelerated creep for the case of heterogeneity in the coefficient of hygroexpansion, but with no moisture gradient. Even though there is no moisture gradient, we still get stress redistribution with each cycle because of the mismatch in hygroexpansion.

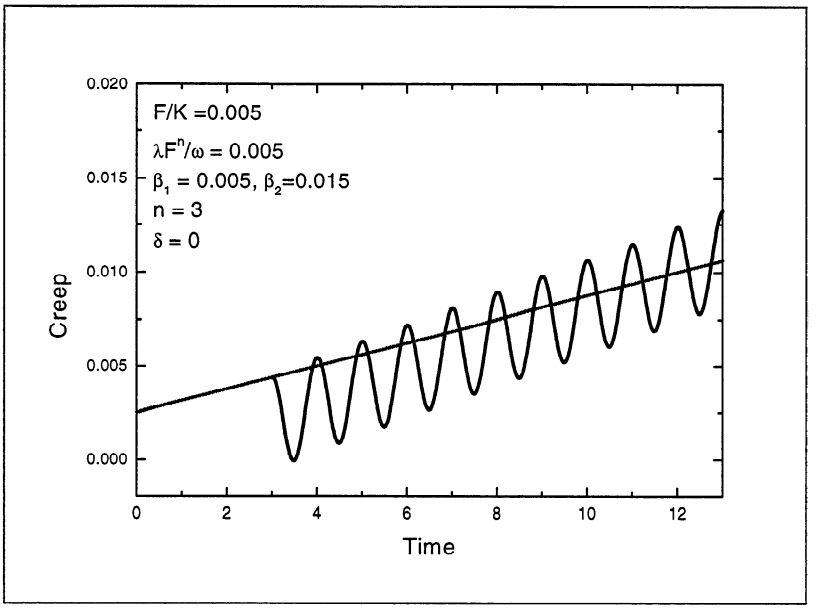

Figure 7. Heterogeneity-driven accelerated creep, with no moisture gradient.

In addition to heterogeneity in hygroexpansion, our model yields heterogeneity-driven accelerated creep if there is heterogeneity in the moisture dependence of material properties. Figure 8 shows accelerated creep as predicted by the model for the case of no moisture gradient and no hygroexpansion, but heterogeneity in the moisture dependence of the spring constants. For this specific model, we increased the nonlinearity to n = 10 in order to get accelerated creep that is clearly visible.

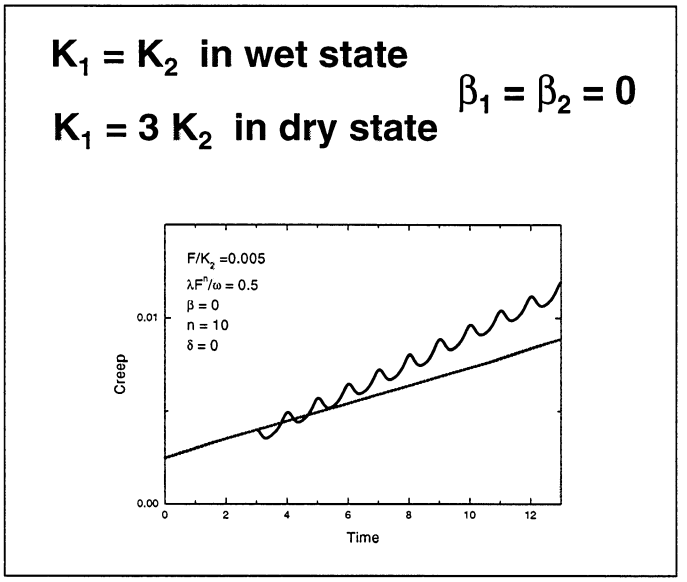

Figure 8. Accelerated creep resulting from heterogeneity in moisture dependence of properties.

Clearly our mechanism produces extra creep. The extra creep can be either moisture gradient driven or heterogeneity driven. The essential ingredients are a creep constitutive behavior that produces excess creep under cyclic loading as compared to the creep caused by application of the mean load, and cyclic redistribution of stresses in the material due to either moisture gradients or material heterogeneity. It remains to establish if this mechanism is the main contributor to accelerated creep in paper. In the next section, we relate some of the predicted behaviors from the model to that observed in paper and similar materials. In Part II of this work we further establish that our mechanism is the likely explanation of accelerated creep in paper.

EXPECTED BEHAVIORS

Based on the proposed mechanism, there are several features about the behavior that stand out and should be observed in experiments. Here we give a few examples that should help establish that the mechanism is active. First and foremost, heterogeneity of material properties is not required for the material to exhibit accelerated creep. We do not require "anisotropic swelling" of the material, we do not require a fibrous material

with bonding, we only need meet the requirements of the mechanism. Thus, a material such as cellophane, which is much more homogenous than paper, should produce accelerated creep if it meets all the criteria. Figure 9 provides the experimental results for a cellophane strip subjected to our accelerated creep test. It is obvious that cellophane exhibits accelerated creep. Numerous repetitions of this test confirm that cellophane exhibits accelerated creep. Moisture-gradient-driven accelerated creep is the most likely explanation for this observation.

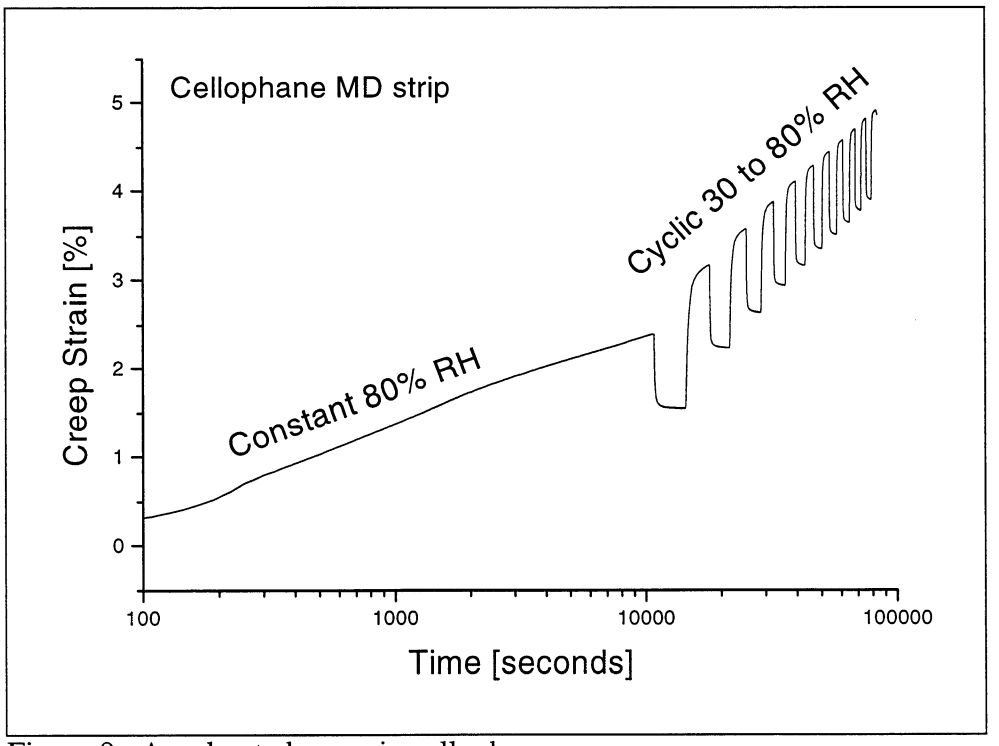

Figure 9. Accelerated creep in cellophane.

Next, our model should conform to what we call the Armstrong test. Armstrong [3] observed that wood did not exhibit accelerated creep under a constant gradient of moisture. This discounted explanations of accelerated creep that relied on the movement of water through the sample. Figure 10 shows the results from our model for the case where only one side of the model is dried and the other side remains wet; clearly, there is no accelerated creep.

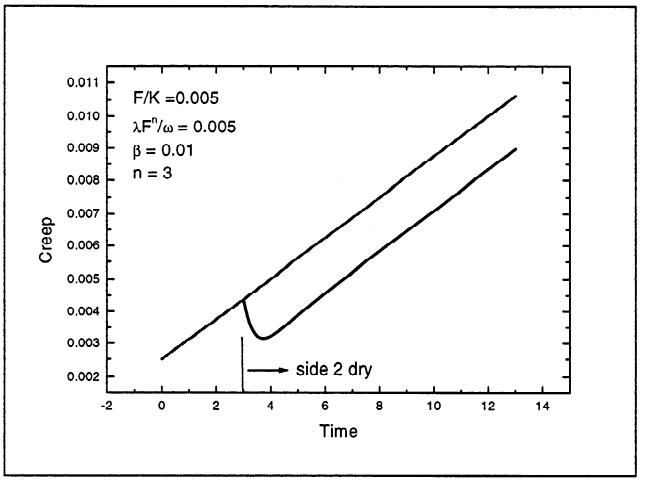

Figure 10. No accelerated creep from constant moisture gradient.

Our model predicts that the accelerated creep ratio should be a function of cycle time. When the cycle time is large compared to the sorption time, the accelerated creep ratio will be small. Of course if the cycle time is shorter than the sorption time, one will not achieve moisture equilibrium and most likely no accelerated creep. Figure 11(a) illustrates the behavior predicted from the model, and Figure 11(b) provides experimental data for TMP paper subjected to different cycle times. Inspection of the figures shows that as the cycle time increases the accelerated creep ratio decreases. We feel that there is an optimum cycle time that would maximize the accelerated creep ratio. Urbanik and Lee [4] reached similar conclusions using a swept sine moisture cycle.

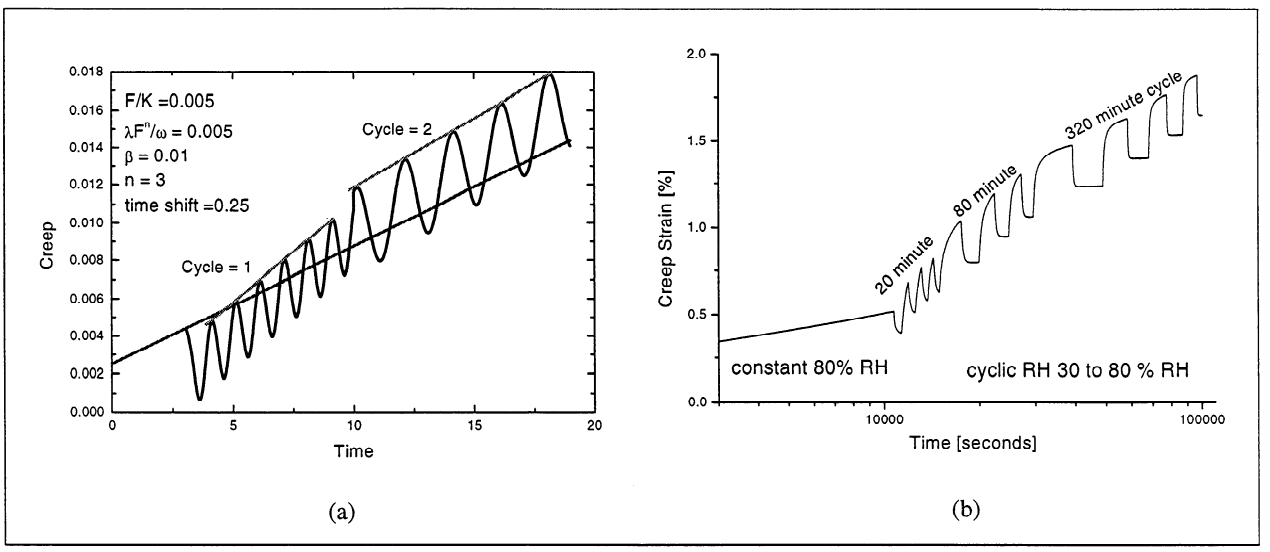

Figure 11. Accelerated creep ratio is a function of cycle time.

We present one final observation. Our model predicts that accelerated creep does not rely on the absolute moisture level in the sheet, but only the magnitude of the change in moisture. Accelerated creep test results for two TMP paper samples cycled between 50 and 10% RH are given in Figure 12. In this case, the accelerated creep ratio is larger than observed for any tests we have completed to date, approximately 13. Note that the creep occurring during the first drying is so severe that it arrests the shrinkage and ends up above the strain that would have occurred if the sample remained at 50% RH. This clearly shows that accelerated creep does not require a certain level of moisture to be initiated.

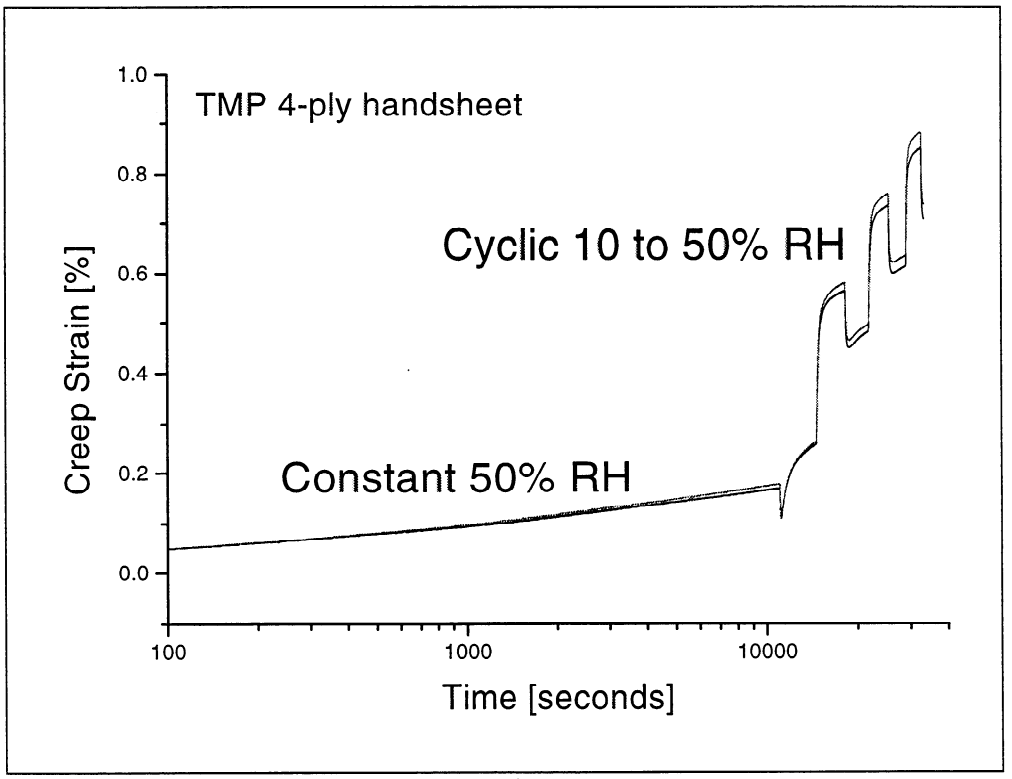

Figure 12. Accelerated creep at low moisture contents.

SUMMARY

Accelerated creep is a natural consequence of the combined effects of the creep, moisture sorption, and hygroexpansive behavior of the material. If the material exhibits more creep upon the application of cyclic load compared to the creep caused by application of the mean load, accelerated creep may occur. For such materials, accelerated creep will occur if either moisture gradients exist for a reasonable time compared to the sorption time or material heterogeneity exists.

Our mechanism maintains that accelerated creep is just creep. If one properly accounts for the creep response of the material as a function of load and history, the sorption behavior of the material, the hygroexpansion, and the material property dependence on moisture content, accelerated creep should naturally fall out of the analysis. In terms of numerical modeling, researchers do not need to make special considerations to predict accelerated creep. The emphasis should be placed on obtaining the correct equations for creep, sorption, and hygroexpansion. To understand accelerated creep in terms of the structure and chemistry of the material, one need not focus on accelerated creep, but focus on understanding creep at constant humidity as well as understanding sorption and hygroexpansion under no load. Our mechanism ties this information together to explain accelerated creep.

Accelerated creep in paper is not as simple as we make it. One still needs to address the apparent interdependence among stress, moisture content, and hygroexpansion. These interactions serve to complicate the situation but we feel they do not alter the basic mechanism present here.

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ACCELERATED CREEP MECHANICS: Part II

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ABSTRACT

In Part I [1], we put forth an explanation of accelerated creep that did not rely on special moisture interactions, new physical phenomena, or unsubstantiated assumptions. Now, we discuss accelerated creep of real materials in terms of the mechanism. Using simple mathematical models, we argue that our accelerated creep criteria are fulfilled in materials that demonstrate accelerated creep (paper, cellophane, and Kevlar fibers) and are not fulfilled in fast sorbing fibers that do not show accelerated creep. We develop empirical creep constitutive equations that fit experimental data for paper and cellophane. Mathematical models, employing these constitutive equations and reasonable assumptions about sorption, produce accelerated creep of the correct form and magnitude. We compare theoretical with experimental results in an attempt to distinguish moisture-gradient-driven accelerated creep from heterogeneity-driven accelerated creep. We explain the lack of accelerated creep observed in some single fibers. Finally, Padanyi's [2] sorption physical aging testing is interpreted as another outcome of the mechanism.

INTRODUCTION

As stated in Part I [1], we believe that accelerated creep is a manifestation of sorption-induced cyclic stress gradients in materials that exhibit more creep when subjected to a variable load than when exposed to a constant mean load. The necessary localized cyclic stresses will be produced in a cyclic humidity environment if significant cyclic moisture gradients develop and/or if the material is heterogeneous in its response to moisture. Accelerated creep follows if the extra creep due to cyclic overcomes the reduced accumulation of creep caused by an average lower moisture content.

RESPONSE TO CYCLIC LOAD

First, we want to demonstrate experimentally that paper and cellophane show extra creep under cyclic loading. Figure 1 provides results of tensile creep tests conducted at three constant load levels and one cyclic load level for a TMP handsheet at 80% RH. The three constant load levels were 15, 25, and 35% of the 50% RH tensile strength. For the cyclic load case, the load was started at the 25% level, cycled between the 35% level and 15% level and finally returned to the 25% level. The average load for the cyclic case was equal to the 25% strength level.

As shown in Figure 1, the cyclic load creep was greater than the creep at a constant 25% load. Thus, we see that this paper meets our constitutive criterion for accelerated creep. Figure 2 documents similar results for an unplasticized cellophane sample loaded in the MD. It exhibited the same behavior.

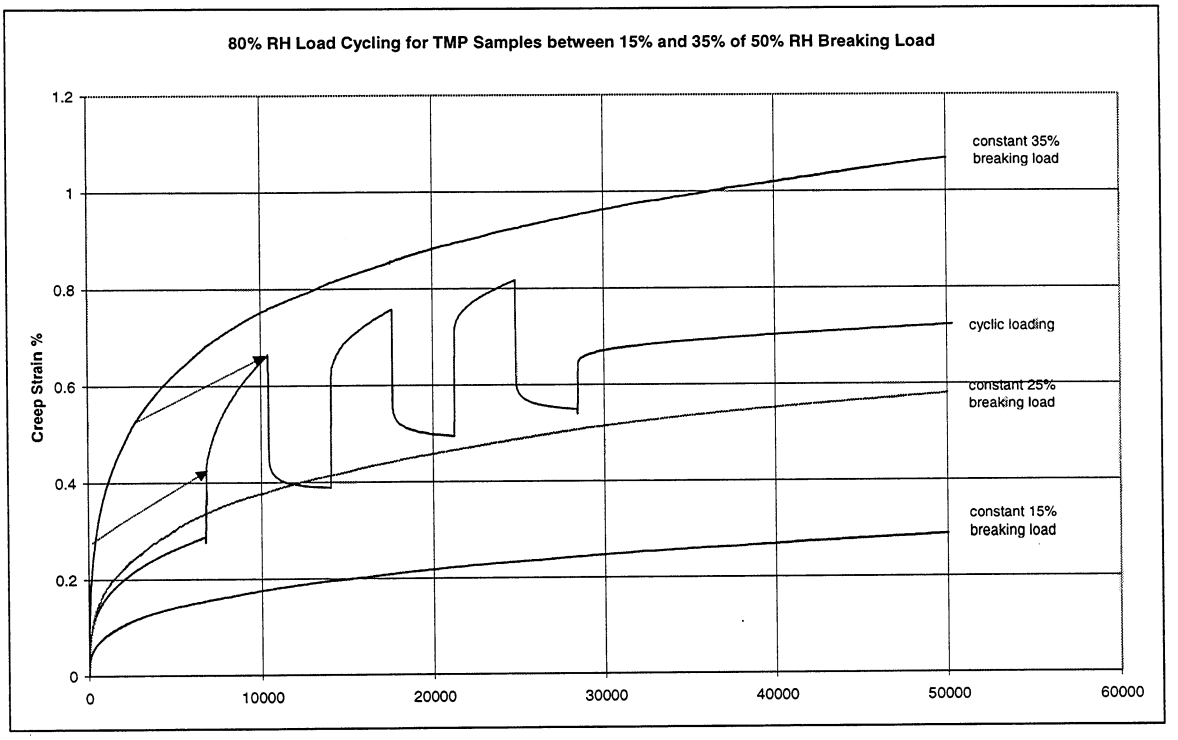

Figure 1. Creep response of paper under cyclic load.

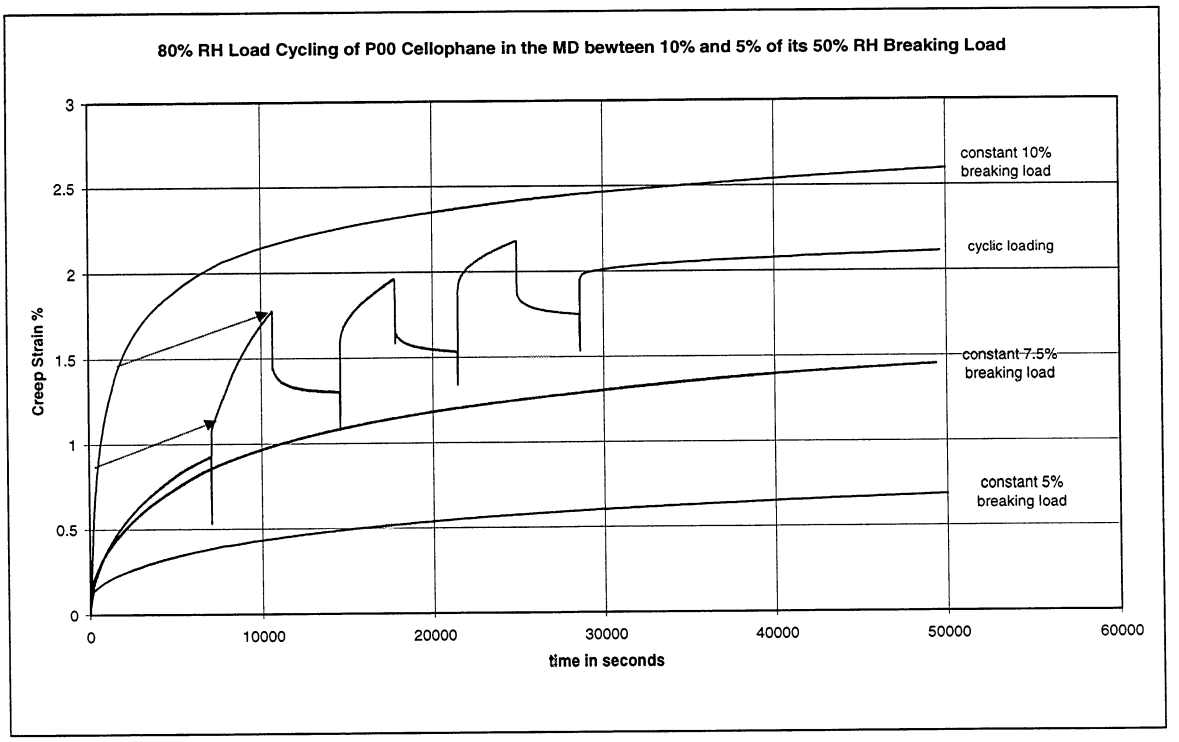

Figure 2. Creep response of cellophane to cyclic load.

From creep curves such as those in Figures 1 and 2, we devised a method to roughly predict creep behavior under variable load from creep curves at constant load. Notice that when the load was first increased from the medium load to the higher level, creep proceeded approximately as it did at the constant high load at the same creep strain. The arrows shown in Figures 1 and 2 demonstrate how the creep curves can be aligned with one another. The vertical offset accounts for the elastic component of the strain that occurred when the load was increased. From this, it looks like the creep rate in a variable load experiment may depend only on the immediate load and creep. Therefore, we propose that variable load creep rates can be constructed, approximately, from constant load creep curves at the same load and creep strain: $d\varepsilon_c/dt = F(\varepsilon_c,\sigma)$. However, this does not account for the creep recovery observed at low-load excursions, and it is not operative when creep and stress are of opposite sign.

MODEL FOR CREEP UNDER VARIBLE LOAD

We incorporated the above observations for creep into a simple expression that was used to predict creep and accelerated creep. From constant load experiments and previous results of Brezinski [3], we know that at long times and high loads paper and cellophane display creep behavior that is linear in log time:

$$\varepsilon_c = K \ln(Ct + 1). \tag{1}$$

We also accepted from Brezinski [3] that a master creep curve could be constructed by a logarithmic time shift proportional to the stress. Then, we wrote a differential equation, which yielded the creep behavior given in Equation (1) for constant load and yielded acceptable behavior for variable load. We further assumed that the total strain was comprised of the creep component, a linear-elastic component, and a hygroexpansive component. The equation governing the time rate of change of strain:

$$\frac{d\varepsilon}{dt} = A\sigma e^{\alpha\sigma-(\varepsilon-\frac{\sigma}{E})/A\sigma} + \frac{1}{E}\frac{d\sigma}{dt} + \frac{d\varepsilon_h}{dt}, \qquad (2)$$

where E is the elastic modulus, ε_h is the hygroexpansive strain, A and B are creep parameters, and α is the master shift coefficient. Equation (2) must not be used for cycling between tension and compression and does not predict creep recovery. It is adequate for tensile creep and tensile creep under a cyclic load. The parameters E, A, B and α are functions of moisture content. The hygroexpansive strain was taken to be proportional to the change in moisture content.

ACCELERATED CREEP

We used Equation (2) on each side of our parallel element model presented in Part I [1]. Next, we applied a tensile creep load and cyclic moisture conditions. We assumed a moisture cycle as shown in Figure 3, where the moisture sorption on one side of our model lags the moisture sorption on the other side by half the sorption time, Ts/2. The moisture cycle shown in Figure 3 starts with both sides of the model in the wet state. The moisture in side one then decreases linearly to the dry moisture content over a time period equal to the sum of half the sorption time and the time it takes for the chamber to reach the dry humidity content, Td. When the element is re-wetted, it takes half the sorption time plus the time required to bring the chamber to the wet humidity conditions, Tw.

We considered two cases: moisture-gradient-driven creep and heterogeneity-driven accelerated creep. All except two of the model parameters were determined from independent tests. The two exceptions are the creep coefficients for the material in the dry state and the amount of heterogeneity from one element to the other. For the moisture-gradient-driven accelerated creep case we assumed no heterogeneity.

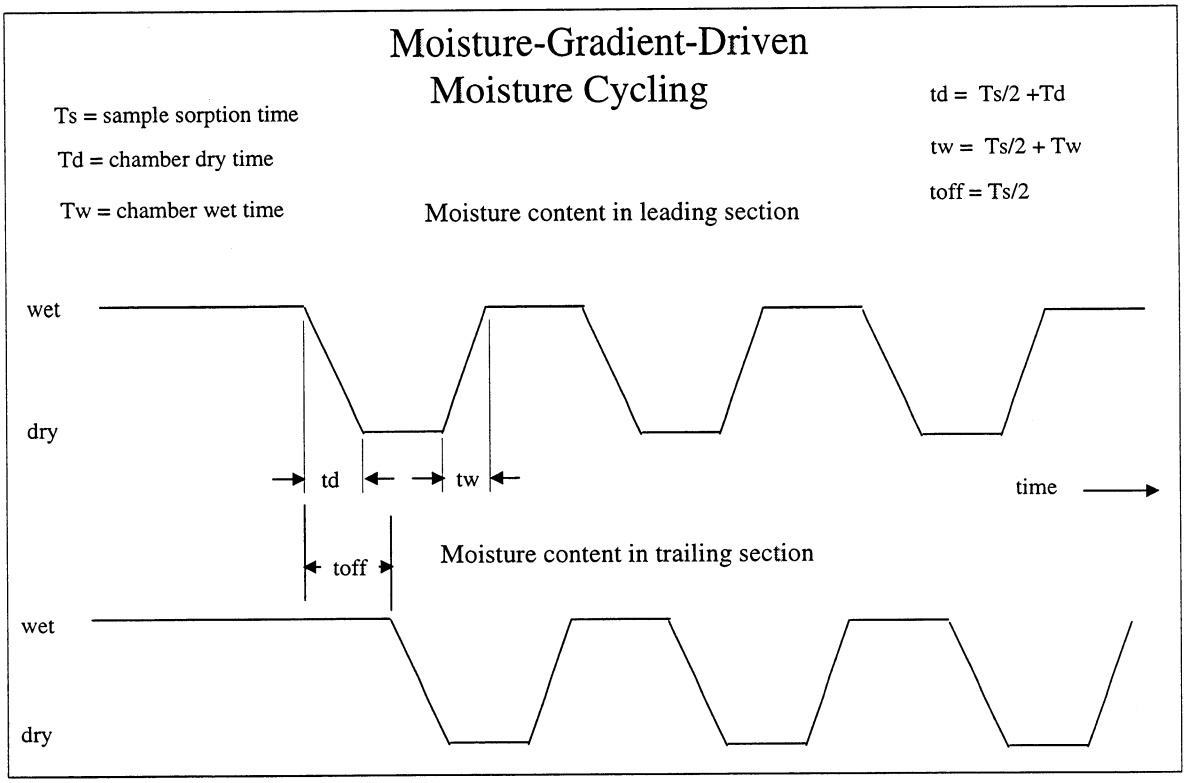

Figure 3. Moisture Profile

A typical accelerated creep curve for paper was given in Figure 1 of Part I. Figure 4 shows the results from the model. Clearly the accelerated creep is similar to that observed in paper. The lower graph in Figure 4 shows how the load on Element 1 of the model varied during the simulation. When the load on Element 1 was high, it experienced larger creep, and when the load on Element 1 was low, Element 2 experienced larger creep.

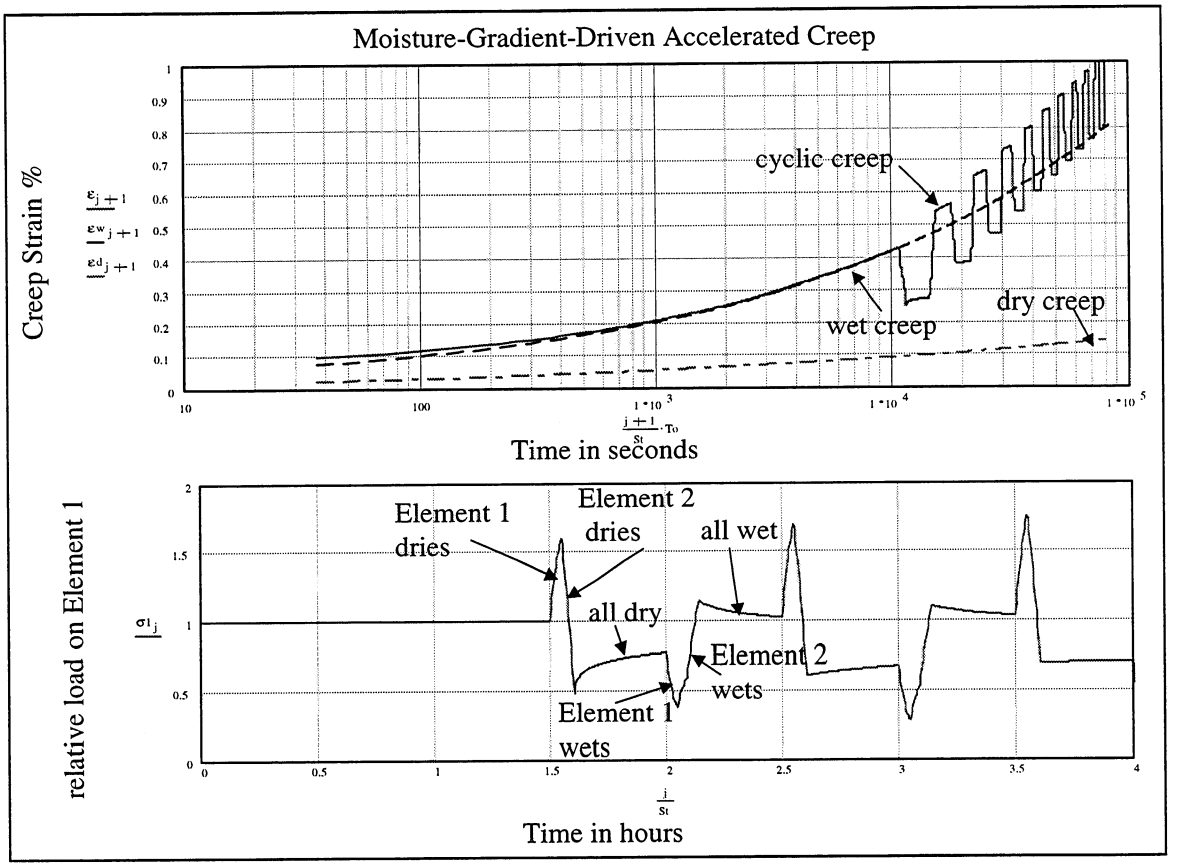

Figure 4. Moisture-Gradient-Driven Accelerated Creep

As shown above, under tensile loading the elements experience their high load exposures when they are dry. Therefore, the mathematical projections of creep rates to high loads and high strains in the dry state are crucial in the prognosis for moisture-gradient-driven accelerated creep. Determination of these from dry, constant-load creep curves would require inordinately long experiments. Thus, we are forced to make an inference. As the projection becomes less aggressive, accelerated creep begins to disappear. Look at Figure 5. This run is a near repeat of the one in Figure 4; only the parameter governing dry state creep projection is reduced.

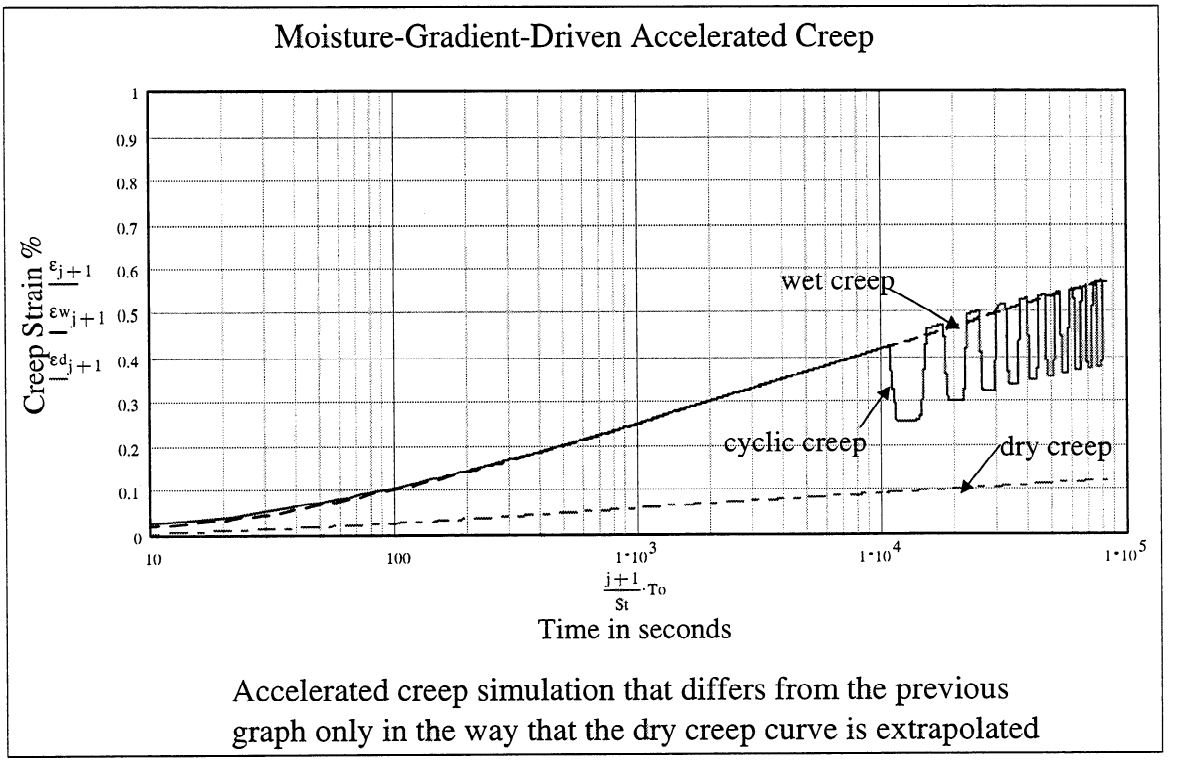

Figure 5. Moisture-gradient-driven accelerated creep with different dry creep parameters.

When we imagine heterogeneity-driven accelerated creep in paper, we picture the sorption-induced load cycling caused by the anisotropy in fiber hygroexpansion. Single fiber hygroexpansion is much greater in the lateral directions than along the axis. The axes of fibers bonded together in a sheet will not, in general, be aligned. Thus, along the direction of tensile load, they will have different coefficients of hygroexpansion. Moisture cycling will induce fiber-level load cycling due to the incompatibilities of hygroexpansion at the bonds. If we assume that the component of hygroexpansion at an angle θ to the fiber axis is proportional to $\sin^2\theta$, we can show that in a sheet of random fiber orientation the average ratio for bonded fibers of the component of hygroexpansion along the tensile axis is about 3.0. Figures 6 and 7 show heterogeneity-driven creep curves using both sets of dry creep curve coefficients. The heterogeneity was produced by assuming a 3 to 1 ratio of hygroexpansion between Element 2 and Element 1, a 1 to 0.75 ratio of elastic modulus, and a 2 to 1 ratio of modulus sensitive to moisture. In this

case, the high-strain dry creep is not so important and accelerated creep is large for both cases. Heterogeneity-driven accelerated creep seems to be more robust in generating accelerated creep from paperlike material properties.

It appears that both moisture-gradient-driven accelerated creep and heterogeneity-driven creep are potent enough to cause accelerated creep in paper. Both types are probably active in paper. It is hard to rationalize a potent heterogeneity-driven mechanism for cellophane. Even though cellophane is more hygroexpansive and more creep prone than paper, it shows less accelerated creep. We attribute this to its reliance on only moisture-gradient-driven accelerated creep.

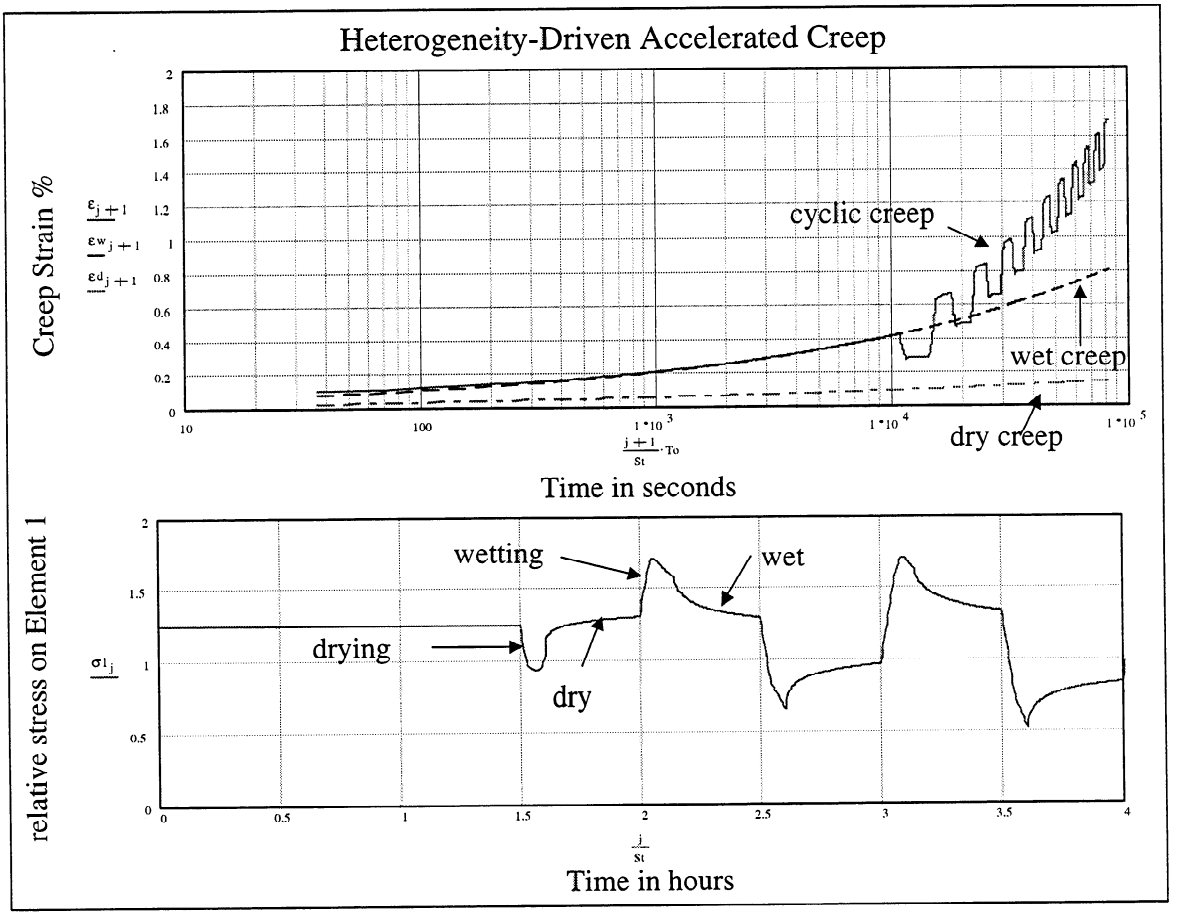

Figure 6. Heterogeneity-driven creep using first dry creep parameters.

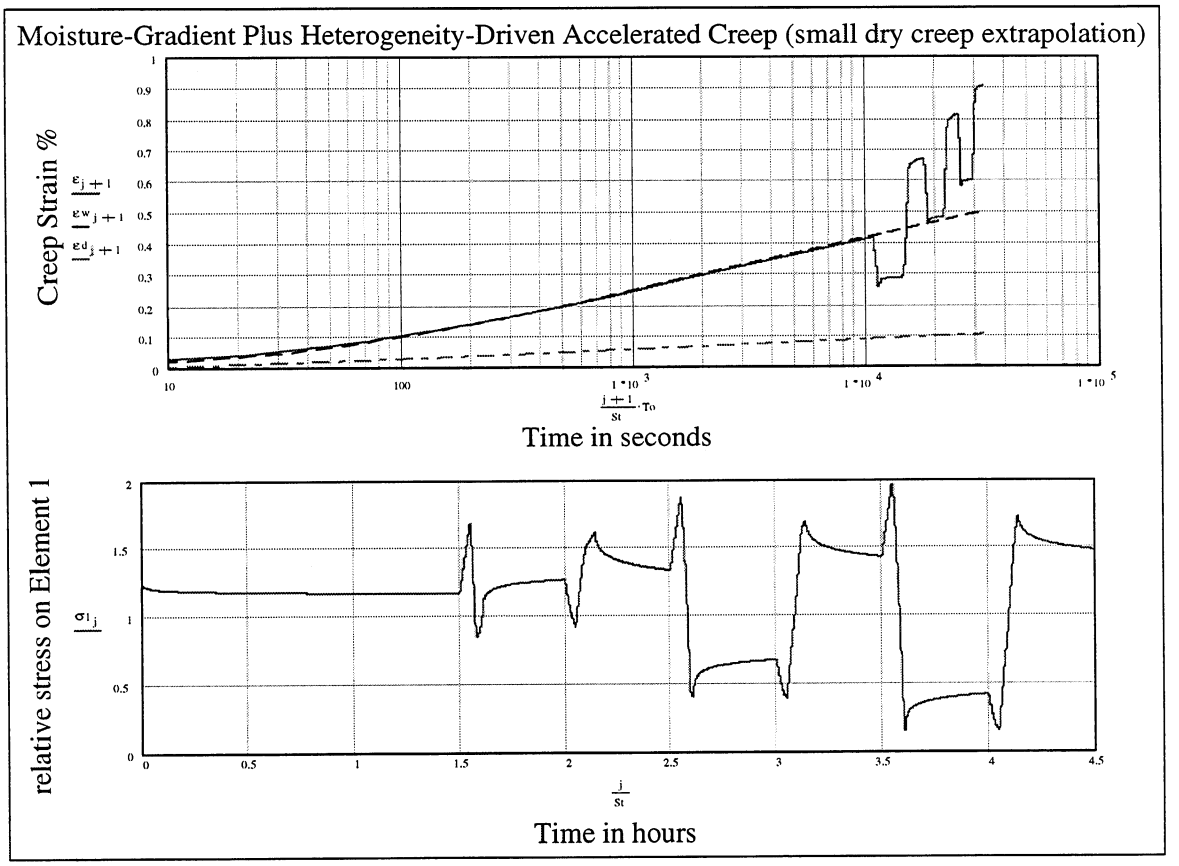

Figure 7. Heterogeneity-driven creep using second set of dry creep parameters.

SINGLE FIBER TESTS

It has been reported that wood pulp, Nylon 6,6, and Rayon single fibers do not exhibit accelerated creep [4,5,6,7], whereas, Kevlar fibers do experience accelerated creep [7]. Figure 8 shows room temperature accelerated creep curves for Kevlar 29 fibers, which we measured in our laboratory. We offer the following explanation for the fiber observations. Heterogeneity-driven accelerated seems unlikely in single fibers; therefore, we consider only the possibility of moisture-gradient-driven accelerated creep in fibers. For the reported tests of single fibers not showing accelerated creep, the time for moisture sorption was small compared to the chamber RH transition times and the humidity cycle times. When a material sorbs more rapidly than the environmental chamber changes state, moisture gradients in the material are greatly attenuated. When material sorption times

are small relative to cycle time, stress concentrations exist for only a small portion of the creep time, reducing the extra creep from stress concentrations. Thus, there is no chance for accelerated creep regardless of the creep constitutive equation.

The sorption times for Kevlar fibers are the order of tens of minutes (9); therefore, stress gradients develop for large portions of the humidity cycles in normal accelerated creep testing. From a sorption time consideration, accelerated creep is now possible. However, to our in initial dismay, creep in Kevlar fibers is insensitive to load [8], and at first glance it appears that Kevlar does not have the creep sensitivity to load necessary to exhibit accelerated creep. Specifically, our constitutive criterion is that a cyclic load causes more creep than a constant mean load. Figure 9 shows that Kevlar 29 does indeed meet this requirement! Thus, even though it has a strange assortment of creep behaviors, Kevlar possesses the combination of creep and sorption properties that qualify it as an accelerated creep candidate.

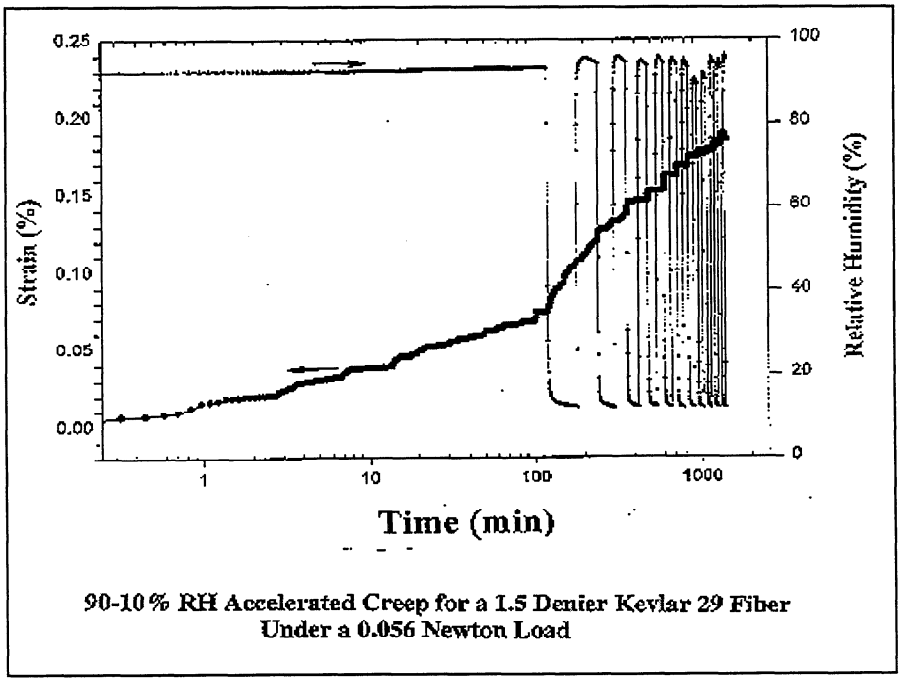

Figure 8. Accelerated creep of Kevlar fibers.

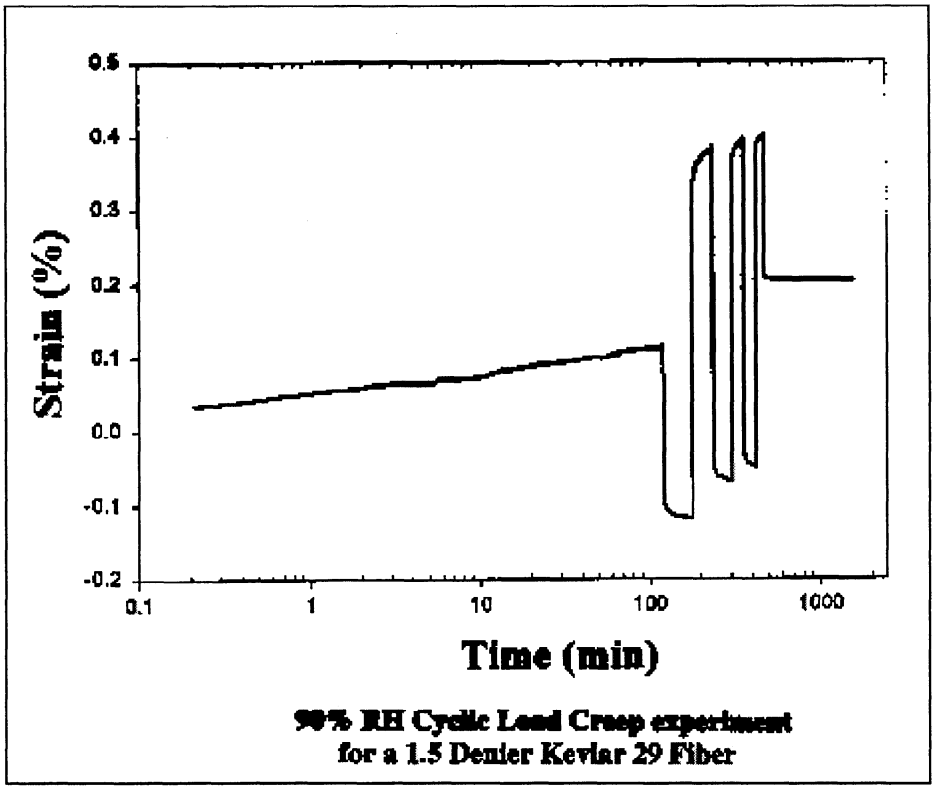

Figure 9. Cyclic load results for Kevlar fiber.

PHYSICAL AGING

The term physical aging is used to describe the apparent stiffening of a material after it has come to thermal equilibrium, but not thermodynamic equilibrium. A polymer that descends from above glass transition to some steady temperature will have a larger creep compliance than the final equilibrium creep compliance for a period of time that far exceeds the time to reach temperature equilibrium. The creep compliance slowly decreases to the final equilibrium compliance. Padanyi [2] observed similar behavior for paper undergoing a moisture change. This could be rationalized for excursions to high moisture since the higher moisture content drops the glass transition temperature. On the other hand, it is difficult to rationalize this explanation for the case where the excursion is to lower moisture contents. As amazing as it seems, Padanyi showed that sending the paper through a low moisture cycle "deaged" the paper. We repeated his experiments and verified his observations.

Padanyi's tests were done at sufficiently high load levels to produce work hardening during creep. Yet, we found that the sample could be repeatedly "deaged" and exhibit, essentially, its virgin creep behavior. If the sample was not "deaged" by drying, we observed work hardening.

We struggled with this for some time until we realized that sorption "deaging" could also be explained in terms of stress concentrations and creep sensitivity to load. When the moisture content of an unloaded sample changes, part of the sample goes into compression and part of the sample goes into tension. This sets up residual stresses in the sheet that relax away slowly after sorption is complete. When the creep load is applied, there will be an uneven distribution of load through the sample. Because of the high sensitivity of creep rate to stress in paper, the extra creep in regions of high stress will overcompensate for the loss of creep in regions of low load. When a piece of paper encounters a sorption event, residual stresses are established making the paper more creep compliant. As time progresses, these residual stresses slowly relax away producing a more creep resistant sample. If, at any time, the sorption event recurs, the sample returns to the more compliant (higher residual stress) state. For us, "deaging" establishes residual stresses, whereas "aging" removes them.

Figures 10 and 11 provide model results (using a hyperbolic sine creep element) of a "deaging-aging" test. Figure 10 depicts the behavior of a sample that is "deaged", "aged", and then loaded and allowed to creep. The bottom curve shows, as a function of time, the load (relative to the load needed to give an elastic strain equal to the hygroexpansive strain) on the element that first responds to the humidity changes. Notice that residual stresses develop during "deaging" and that they slowly dissipate during "aging".

The graphs of Figure 11 represent just the creep portion of model experiments. The solid curve in the top graph is the creep curve of a material "aged" for 100 time periods, and the dashed line represents a very old sample. In the bottom curve the solid line

represents the creep response in the material after "aging" for only 1 time period, and the dashed line is the same as in the top graph. Clearly the sample that has been "aged" the least shows the greatest creep.

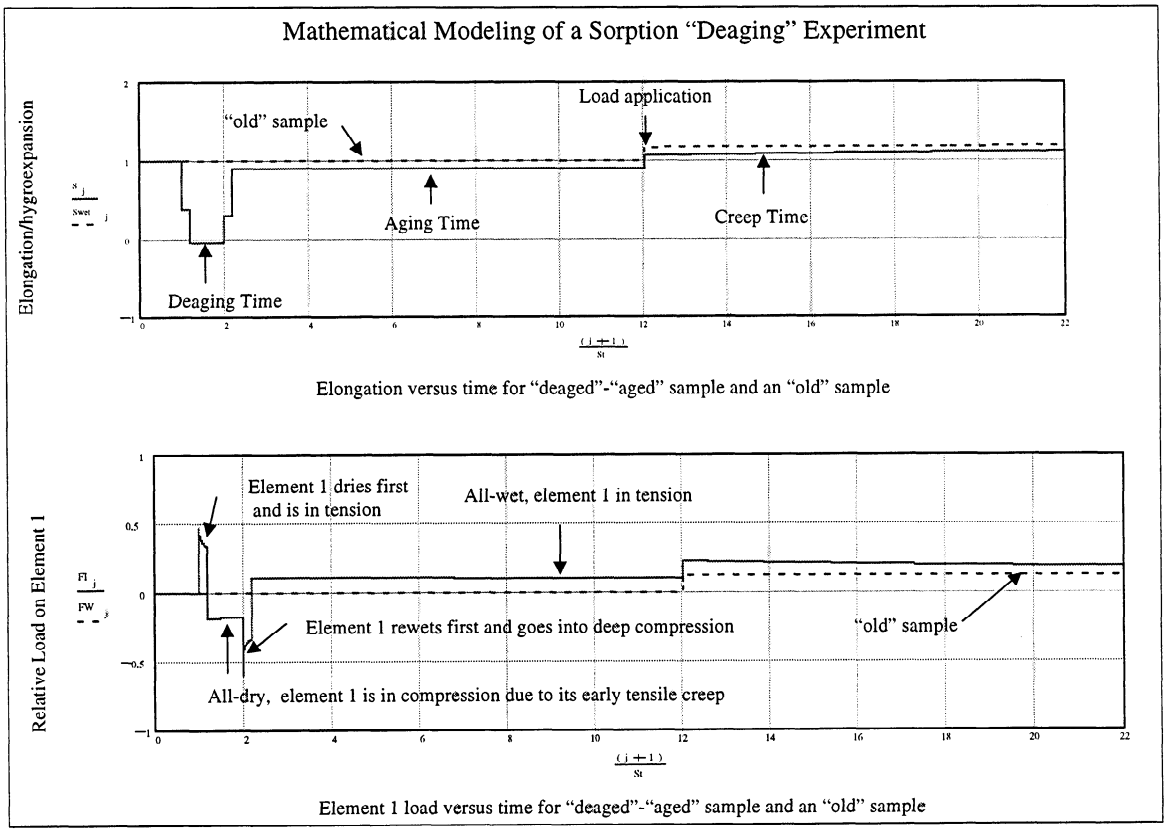

Figure 10. Stresses during de-aging and aging test.

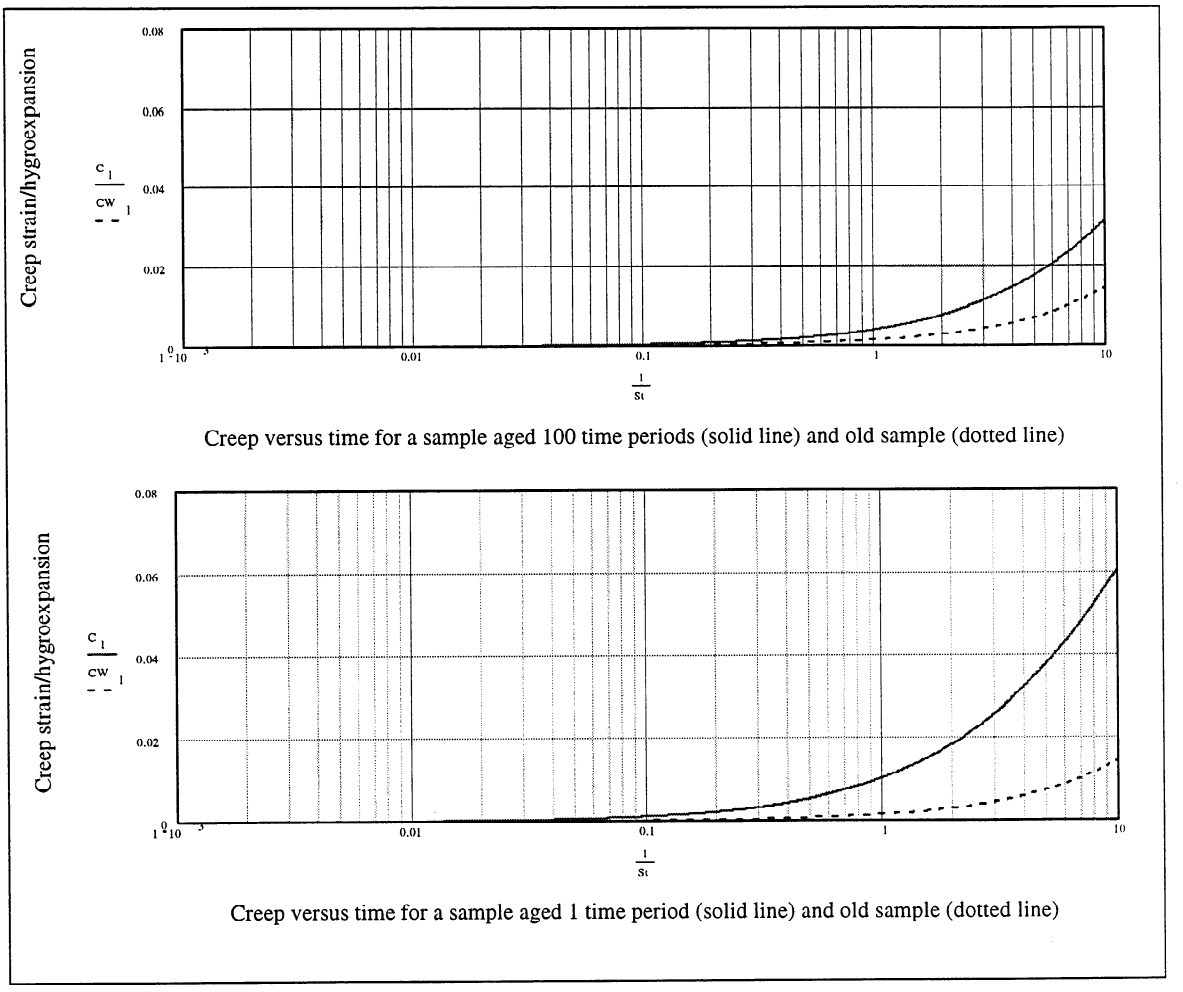

Figure 11. Effect of "aging" on creep response.

CONCLUSIONS

By developing a creep equation that adequately represents the creep behavior of paper under variable load, we have shown that the mechanism presented in Part I [1] is most likely active in paper. Cyclic stress gradients caused by moisture gradients or heterogeneity combined with paper's response to cyclic loads lead to accelerated creep. A comparison of our simulated creep curves with paper experimental data convinces us that heterogeneity-driven accelerated creep is active. Moisture-gradient-driven accelerated creep may play a role as well. We feel we have adequately demonstrated the reasons why (in contrast to Kevlar fibers) pulp fibers do not exhibit accelerated creep.

Finally, we argued that paper's high sensitivity of creep to load is the cause of sorption "physical aging" as observed by Padanyi.

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